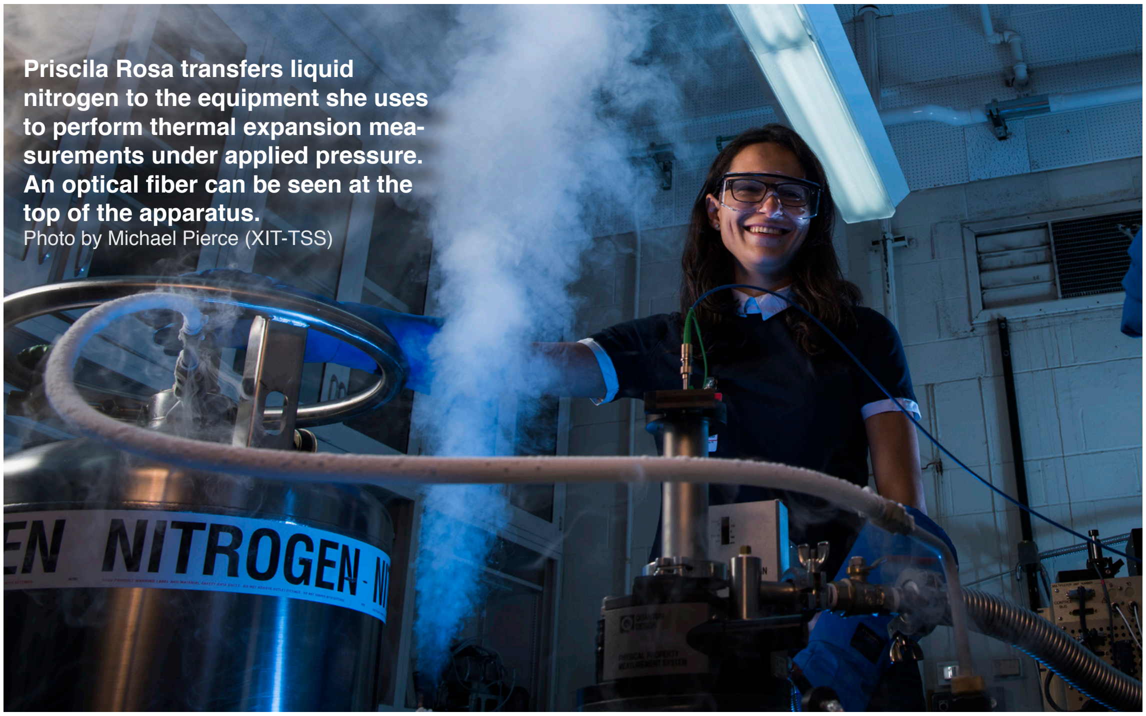
June 2018
Approved for public release;
distribution is unlimited.

MPA Materials Matter

INSIDE



Priscila Rosa transfers liquid nitrogen to the equipment she uses to perform thermal expansion measurements under applied pressure. An optical fiber can be seen at the top of the apparatus.
Photo by Michael Pierce (XIT-TSS)

Priscila Rosa

Putting the pressure on to expand scientific possibilities

By H. Kris Fronzak, ADEPS Communications

> *The knowledge gained from fundamental research is priceless.*

Priscila Rosa knows the importance of applying pressure to achieve goals. That's how she summoned the drive to leave Brazil, her home country, for a joint postdoctoral fellowship in the United States.

Now a materials physicist in Condensed Matter and Magnet Science (MPA-CMMS), Rosa applies pressure to understand how materials behave in extreme environments. She is the principal investigator of an early-career Laboratory Directed Research and Development (LDRD) project using pressure to measure thermal expansion in quantum materials. This fundamental understanding is essential to ultimately knowing how to control and tailor such materials for potential applications.

The LDRD project satisfies a personal goal of Rosa's to explain how and why things work. "The knowledge gained from fundamental research is priceless. It allows you to understand things so well that you can predict and control phenomena," she said. "I'm at the Lab because I believe in the power of fundamental research."

continued on page 4

From Tanja's desk . . .

"On behalf of the Division Office, I would like to warmly welcome all students and wish you a fantastic summer!"

Summer is upon us and change is on the horizon! On June 8 the new contract was announced. By the time you read this note, we will hopefully know more what this means for MPA. In the meanwhile, let's focus on what MPA does best, which is outstanding science. As most of you know by now, Rick is taking some well-deserved time off and I'm happy to welcome Brad Beck as our acting deputy division leader. Brad's been with the Lab for 26 years and has broad experience in GS and NW. He is a nuclear engineer by training, so we are retaining the engineering scheme (as you may know, Rick is a mechanical engineer). Please welcome Brad on board! He'll be right here in the Division Office in 03-1415, the building behind CINT.

This is Susie's favorite time of the year—our summer students are finally here! Many thanks to Vanessa, who for the third year serves as the division's student liaison. On behalf of the Division Office, I would like to warmly welcome all students and wish you a fantastic summer! If there is anything we can do to help you be happier or more successful, please talk to your mentor, group leader, or us in the Division Office. Be on the lookout for the many opportunities that LANL offers our summer students, especially those that allow getting to know what else is going on in the Lab.

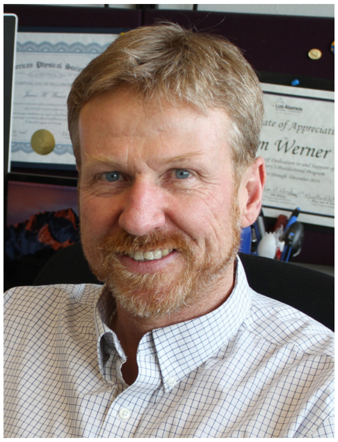
We would love to have a logo for our division, and we hope that you will help us with your creativity! I'm a basket case when it comes to art, so we really need some help. Our goal is to provide you with opportunities to learn about the other organizations in MPA so that you can come up with a logo that represents all of us. For more details about this, contact Nicole Strother in MPA-CMMS.

On a safety note, the fire danger is extremely high and it is important that everyone does their part to protect our community. This means abiding by all closures and making sure that we know how to reach you should the worst happen and we need to evacuate.

Please enjoy the beautiful weather, have a wonderful vacation (if you are so lucky!), and have a great summer. It's also my favorite time of the year!

MPA Division Leader Tanja Pietraß

Tanja



From Jim's desk . . .

“ ‘— *acting deputy—who absolutely needs to get in touch with that guy!*’ ”

For my initial “From the desk” piece, I figured I’d share my thoughts about the transition from being a scientist to being a manager. For those who don’t know me, I’ve been at LANL about 20 years (~3 as a postdoc, ~16 as a staff scientist, and ~1.5 in management). As a scientist, I primarily worked (and still try to work) in developing new tools and instrumentation for biophysical measurements on the nanoscale, with an emphasis on single molecule detection by laser-induced fluorescence.

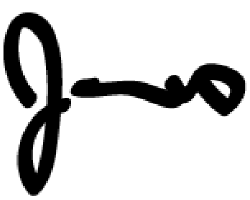
As far as my LANL management experience is concerned, over the past year and a half I have been an acting co-deputy group leader, an acting deputy group leader, an acting group leader, and a “permanent” deputy group leader. As the observant reader has surmised by now, I have experienced a few rungs on the management career ladder and can give people my views on the pros and cons for a couple of these rungs. As I told one of my friends, being acting group leader was a lot harder than being acting deputy. This person then said, “I can see that—*acting deputy*—who absolutely needs to get in touch with that guy!”—a phrase that still brings a smile to my face.

Seriously, though, as far as the transition into management goes, I think the closest comparison most scientists can relate to is the transition from being a postdoc to being a technical staff member. During my salad days as a postdoc at the Lab, I was allowed to spend close to 100% of my time on science. When I became a staff scientist and realized how much my mentors had done to protect me (e.g., securing funding, taking care of safety documentation, taking care of chemical disposal paperwork), I thanked my former mentors (Peter Goodwin, Dick Keller, and Brian Dyer) as I had no clue how many bureaucratic tasks they shielded me from. I feel the same way now as a manager. I want to thank those who came before me and current managers for doing what they can to make life for the scientists and others at the Lab easier.

I can say that life as a manager is never dull. The days fly by faster than you can imagine. I used to think I was pretty busy as a scientist. As a scientist, it always seemed to me like I had a bunch of papers to write, a bunch of proposals to write, a bunch of papers to review, and a bunch of proposals to review, all while still trying to spend some time in the lab to do experiments. I can say now, though, after being in management for a while, I have a new appreciation for how much free time I actually had as a scientist.

While it does demand a lot of time, being a manager can be quite rewarding. When I first started my acting management role, I was taken aback by the fact I would sometimes get “thanked” for doing things that seemed to me to be a routine part of the job. I wanted to say (and sometimes did), “No need to thank me dude, you know they are paying me for this.” While this statement about being paid is true, I do appreciate everyone who has thanked me for trying to do my best here in this new role.

So, by now, if you are still reading this, I am sure you must want to try your hand at management. I have good news for you. Andreas and I are more than willing to schedule simultaneous extended vacations so you can take on an acting role for a prolonged period. Just let us know when you’d like to begin your personal management journey.



CINT Deputy Group Leader Jim Werner

Rosa cont. ...

Rosa joined a group perfect for the task. MPA-CMMS has a distinguished record of supporting the Lab's national security science mission through fundamental materials discoveries. These contributions started with post-World War II experiments that determined the properties of the newly created element plutonium. They continue with recent breakthroughs such as the discovery of the first plutonium-based unconventional superconductor, the detection of the long-sought nuclear magnetic resonance signature of plutonium-239, and the first-ever resolution of delta plutonium's electronic ground state.

As a member of the group's Strongly Correlated Electron Systems team, Rosa synthesizes single crystals—a delicate process crucial to understanding a material's basic properties—and explores those crystals under pressure. She focuses on correlated electronic systems based on *f* electrons, a renowned area of leadership for Los Alamos. Electrons in conventional metals and semiconductors, e.g., copper and silicon, act independently. In correlated materials containing *d*- or *f*-electron incomplete shells, about 10^{23} electrons interact strongly with each other, creating complex, emergent ground states—plutonium being a classic example.

"Priscila is unique in the sense that she works in two realms: sample synthesis and characterization," said Mike Hundley, MPA-CMMS group leader. "She's built up great intuition for sample growth and for what combinations of elements could lead to materials that support new discoveries."

Rosa is the principal investigator for two LDRD projects and participates in two other Department of Energy projects, all aimed at investigating quantum materials—a class of materials that promise to revolutionize energy- and security-relevant technologies.

To do this she has developed novel experimental techniques and diagnostics tools. For example, her first project as a Director's Postdoctoral Fellow resulted in a general principle for using hydrostatic pressure to force cerium-based superconductors to nucleate a magnetic phase. Last year she proved that optical fiber sensors can be used to measure the thermal expansion of these superconductors under pressure. This has led to her early-career LDRD project that uses applied pressure as a "tuning knob" to define the thermal expansion of the strongly correlated superconductor $CeRhIn_5$ at the point with the largest quantum fluctuations.

"Priscila's foundational research has a common theme of discovering and understanding emergent quantum states of matter. It pushes the forefront of science and opens possibilities for new, quantum-based technologies," said her mentor Joe Thompson (MPA-CMMS).

To further expand her contributions to the Lab's energy security efforts, she recently submitted an LDRD proposal with Jinkyoung Yoo (Center for Integrated Nanotechnologies, MPA-CINT) to investigate *f*-based nanowires, which are predicted to host unusual particles that enable quantum computing. By measuring the electrical properties of these strongly correlated nanowires, the team aims to detect these particles for the first time in an f-electron-based material.

"Priscila has a particularly curious mind which is reflected, in part, by the breadth of research that she leads," said Thompson. "She is committed to the success of her peers, the organization, and her own work, no matter how daunting the challenges."

Priscila Rosa's favorite experiment

What: Thermal expansion measurements under pressure using optical fibers.

Why: To realize an unprecedented test of quantum theories.

When: 2017

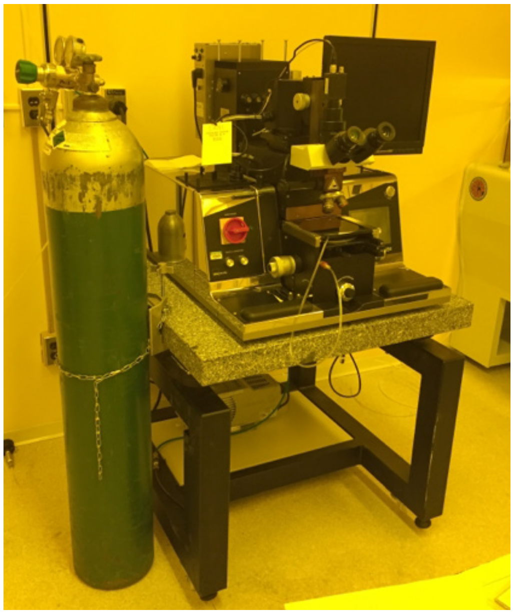
Where: Los Alamos National Laboratory

Who: Priscila Rosa and Sean Thomas (Condensed Matter and Magnet Science, MPA-CMMS), Fedor Balakirev and Jon Betts (National High Magnetic Field Laboratory, NHMFL-PFF); Soonbeom Seo, Eric Bauer, and Joe Thompson (MPA-CMMS); and Marcelo Jaime (NHMFL-PFF).

How: Using optical sensors in a high-pressure environment by feeding an optical fiber inside the sample chamber of a pressure cell.

The "a-ha" moment: The most significant challenge of this experiment was that the optical fibers kept breaking at the boundary between the low- and the high-pressure environments. The "a-ha" moment was to determine, after a conversation with Adam Dioguardi (formerly MPA-CMMS), that we could coat the fibers with a metallic layer to prevent breakage. This change resulted in an important new capability for researchers studying correlated electron materials.

Newly upgraded, well-equipped cleanroom available for unclassified R&D



The cleanroom includes a Karl Suss MJB3 mask aligner for photolithography. This system can accommodate photomasks up to four inches and creates patterns with a resolution of 500 nm.

Newly upgraded, unclassified, certified ISO class 3 and class 4 cleanrooms, equipped with state-of-the-art micro- and nano-fabrication equipment, are available at the Laboratory.

Operated by the Materials Physics and Applications (MPA) Division, the facility caters to research and development that calls for a clean, controlled environment. Located in Technical Area 3, SM-40, which is in the open security environment, the facility offers easy access and a variety of tools to accommodate diverse research needs, from a mask aligner (see photo, below) to vapor deposition to a wire bonder and, soon, systems for reactive ion etching and plasma-assisted atomic layer deposition.

Key to a cleanroom's operation is the filtration system, which recirculates air, minimizing potential contaminants that could prove detrimental during device preparation. The new class 3 cleanroom has fewer than 1,000 particles larger than 0.1 μm per cubic meter, while the two ISO class 4 rooms can have a maximum of such 10,000 particles per cubic meter. In comparison, the air we breathe may have as many as 35,000,000 particles per cubic meter that are 0.5 μm or larger. The facility is not bound to a specific certification process or designed for a particular research topic, which is especially advantageous to one-off users who want to research and develop devices. The facility's flexible nature and new equipment make it useful for Los Alamos's programmatic needs, including prototype device fabrication at small scales.

Materials Synthesis and Integrated Devices (MPA-11) Group Leader Andrew Dattelbaum headed the facility's refurbishment along with MPA staff Abul Azad and Jinkyoung Yoo (both Center for Integrated Nanotechnologies, MPA-CINT) and Chris Romero (MPA-11), who provided equipment acquisition and installation support.

Chris Romero (MPA-11) is the cleanroom point of contact for daily operations and will help organize scheduling, training, and work execution for interested users.

Romero is point of contact for daily operations and will help organize scheduling, training, and work execution for interested users. CINT, a DOE Office of Science User Facility, is helping support the cleanroom facility at this time. In particular, scientists interested in using the facility can potentially get free access to its capabilities through the CINT user program. Contact Azad for more information about accessing the facility through CINT.

The cleanroom's revamp was funded by institutional support from Los Alamos National Laboratory.

Technical contact: Andrew Dattelbaum

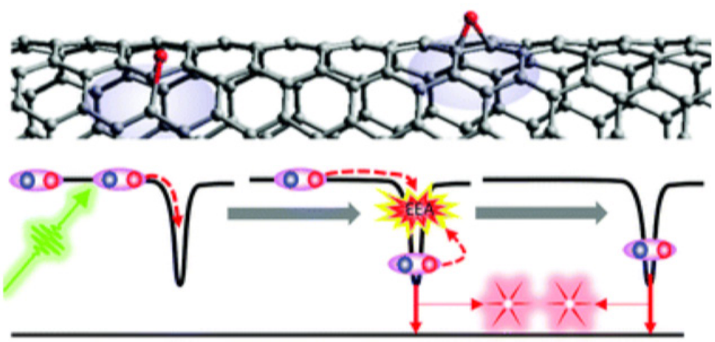
Multi-excitons of doped nanotubes show new path to lasers and entangled photon sources

Doping of single-wall carbon nanotubes via covalent attachment of chemical functional groups has rapidly emerged as an effective way to enhance these nanotubes' emissive properties and introduce new functionalities and has established solitary dopant states as a new type of quantum light source. Specifically, improving emission efficiency brings new promise for single-wall carbon nanotube-based bioimaging, light-emitting devices, and lasers.

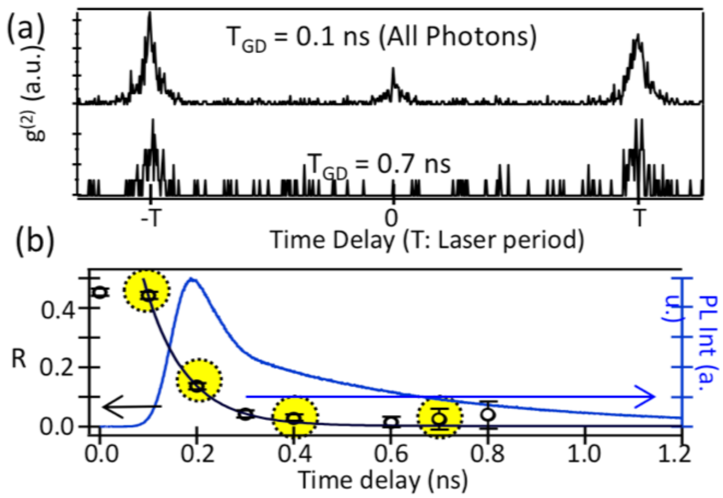
While recent studies have brought new understanding of the electronic structure, non-linear optical properties, and photoluminescence fluctuation behaviors of doped nanotubes, many critical photophysical processes remain unclear. Using a novel experimental approach and a theoretical kinetic model, Los Alamos researchers shed light on the rich multi-exciton processes associated with the dopant states, explaining key experimental observations. Their work, conducted using single nanostructure optical spectroscopy and carbon nanotube functionalization capabilities at the Center for Integrated Nanotechnologies (CINT), opens a new path toward carbon nanotube-based lasers and entangled photon pair sources.

They showed that, while most dopant states emit one photon per one excitation cycle and can therefore serve as single photon emitters, some dopant states emit photons in pairs. These photon pairs can come from two dopant states located within the laser excitation spot or from successive recombination of two excitons (electron-hole pairs) in a single defect. By performing a time-gated, second-order photon correlation experiment, the researchers demonstrated that the photon pair emission originated from two successive

continued on next page



Capture and recombination of two excitons (blue and red spheres bound together) by oxygen dopants (top, red balls) of a carbon nanotube (top) lead to emission of photon pairs (bottom pink stars).



A time-gated $g^{(2)}$ experiment separates photons from the fast and slow decays of multi- and single excitons. Before separation, $g^{(2)}$ shows a peak at zero time delay representing efficient multi-exciton emission (a: top trace). The center peak decays with increase of gate time (b) and becomes zero after 0.7 ns of gate delay (a: bottom trace).

Multi-excitons cont.

capture and recombinations of excitons by a single dopant state. Further experimental and theoretical analysis showed that this type of photon pair emission process can have a 44% efficacy of the single photon emission and that the main limiting factor is annihilation of excitons upon collision at high excitation power.

This work, conducted at CINT, a DOE Office of Basic Energy Sciences user facility jointly operated by Sandia National Laboratories and Los Alamos National Laboratory, was supported by Los Alamos's Laboratory Directed Research and Development program. It supports the Laboratory's Energy Security mission area and its Materials for the Future science pillar, particularly its emergent phenomena science theme by advancing understanding of how to tailor a material to perform in ways beyond its basic properties, enabling both controlled functionality and predictable performance, the core vision of the Laboratory's materials strategy.

Researchers: Xuedan Ma (formerly MPA-CINT, now Argonne National Laboratory); Nicolai F. Hartmann (formerly MPA-CINT, now neaspec GmbH in Munich); Kirill A. Velizhanin and Sergei Tretiak (Physics and Chemistry of Materials, T-1), Jon. K. S. Baldwin, Stephen K. Doorn, and Han Htoon (MPA-CINT); and Lyudmyla Adamska (formerly MPA-CINT, now Boston University). Reference: "Multi-exciton emission from solitary dopant states of carbon nanotubes." *Nanoscale* **9**, 16143 (2017).

Technical contact: Han Htoon

Exploring materials in extremes with novel sensor techniques

Measuring thermal expansion and magnetostriction under hydrostatic pressures

Thermal expansion measurements of materials provide critical insights for a wide variety of applications, ranging from materials engineering to biological system simulations to studies of thermodynamic phenomena such as phase transitions. However, precisely determining thermal expansion under hydrostatic pressure is hindered by the small working volumes typical of pressure cells, which make the standard capacitance dilatometry approach impossible.

In a study published in *Sensors*, Materials Physics and Applications researchers report the first implementation of thermal expansion and magnetorestriction measurements using fiber Bragg grating (FBG) sensors at cryogenic temperatures and hydrostatic pressures of 2.0 GPa. FBG measurements under pressure have been reported, but only within the MPa range. Their results reveal the advantages and the limitations of this technique in the GPa region and open an unexplored route to the understanding of materials under pressure.

The researchers took an optical approach to determining the thermal expansion of materials under hydrostatic pressure. The approach uses FBGs as strain sensors inside of a pressure chamber to measure strain in the antiferromagnetic material $CeRhIn_5$. $CeRhIn_5$ is a highly tunable, exceptionally impurity-free antiferromagnetic metal that turns into an unconventional superconductor under applied pressure. Because of this, $CeRhIn_5$ has become a prototype of quantum criticality and of quantum fluctuations that arise under pressure. Determining the thermal expansion of this system under pressure could lead to long-sought predictive understanding of quantum criticality in these systems. The determination could also mean new opportunities for exploiting quantum fluctuations and the self-organizing states.
At high temperatures, temperature compensation was hindered by the freezing point of the chosen pressure transmitting medium. However, temperature effects at low

continued on next page

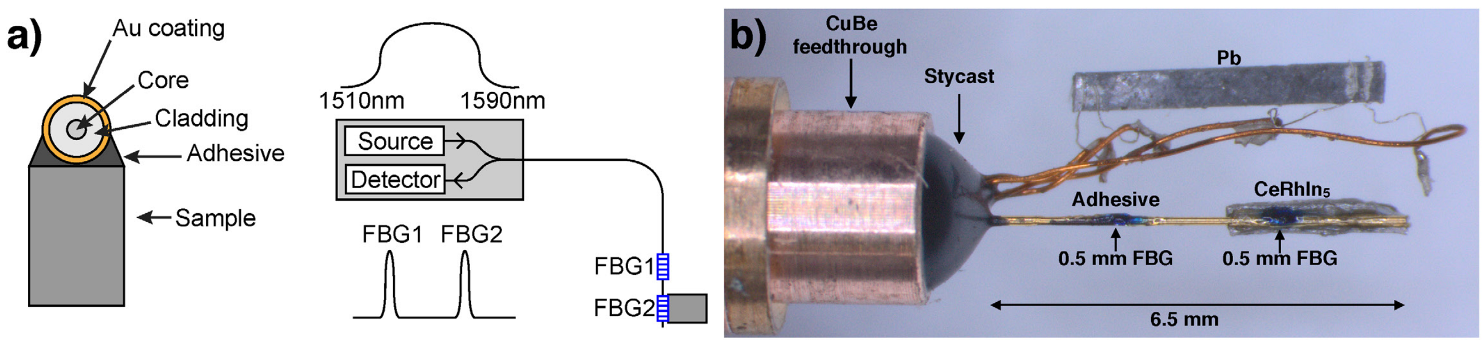
Novel sensor techniques cont.

temperatures were negligible, which enabled the team to simultaneously measure two Bragg gratings using an optical sensing instrument capable of resolving strains in the order of dL/L = 10-7, i.e., changes in length of the order of an Angstrom. The results showed that high-resolution thermal expansion measurements can be performed under pressure, which opens new avenues of research for a broad range of materials.

The work supports the Laboratory's Energy Security mission area and its Materials for the Future science pillar by exploring ways to create materials with controlled functionality and predictive performance, the core vision of the Lab's materials strategy. It was sponsored by the NHMFL User Collaboration Grant Program, the DOE Basic Energy Sciences' "Complex Electronic Materials" and "Science at 100T" programs, and Los Alamos' Institute for Materials Science. The study was partially performed at the National High Magnetic Field Laboratory, which is supported by National Science Foundation, the state of Florida, and the DOE.

Researchers: Priscila Rosa and Sean Thomas (Condensed Matter and Magnet Science, MPA-CMMS), Fedor Balakirev and Jon Betts (National High Magnetic Field Laboratory, NHMFL-PFF); Soonbeom Seo, Eric Bauer, and Joe Thompson (Condensed Matter and Magnet Science, MPA-CMMS); and Marcelo Jaime (NHMFL-PFF). Reference: "An FBG optical approach to thermal expansion measurements under hydrostatic pressure," *Sensors*, **17,** 2543 (2017).

Technical contact: Priscila Rosa



The experimental setup for pressure-dependent measurements.

Experiments expand fiber Bragg techniques for dilatometry studies in extreme magnetic environments

The crystal lattice of materials responds to changes in the environment, including magnetic and electric fields and chemical environment. Techniques that can detect such changes with a small footprint and high sensitivity have applications in multiple fronts. In an article published in *Sensors*, researchers at Los Alamos and their international collaborators discussed the development of a high-sensitivity optical interferometry-based dilatometer, i.e., a method to detect changes in lattice parameters as small as one part in 100 million. Such a method would reduce the timescale from milliseconds to nanoseconds for measurements and could be used in some of the most extreme sample environments.

The team experimented with three different approaches to single-mode SiO_2 fiber Bragg techniques for dilatometry studies on small, single crystalline samples in high-magnetic fields and low temperatures. Fiber Bragg gratings (FBGs), generally speaking, are appropriate for sensing in extreme conditions due in part to their immunity to electromagnetic interference and mechanical vibrations. The three specific FBG approaches were chosen on the basis of the data acquisition speed that make them suitable for experiments in magnetic fields of different durations.

One approach used a swept wavelength laser source to interrogate FBGs when magnetic fields are produced in a continuous fashion. This approach achieved a strain resolution approaching $\Delta L/L \approx 10^{-8}$. A second 46-kHz setup based on an InGaAs line array camera was used in cases where the sample was exposed to millisecond-long magnetic fields. This method resulted in typical resolutions of $\Delta L/L \approx 10^{-7}$–$10^{-5}$, depending on the type of magnet. Finally, a time-delay dispersive pulsed laser approach was implemented for the most demanding sample environments in microsecond-long magnetic field pulses produced by a single-turn type magnet. The strain sensitivity achieved by this final approach was better than 10^{-4}, the highest sensitivity ever achieved anywhere in the nanosecond timescale.

The study also examined typical shortcomings and artifacts when measuring millimeter-long samples in pulsed magnetic fields at cryogenic temperatures and completed a quantitative evaluation of existing analytic models for strain-lag across sample/optical fiber interfaces. These examinations showed that small samples in the millimeter range performed better than predicted by available models. The team is now conducting complementary studies to identify strate-

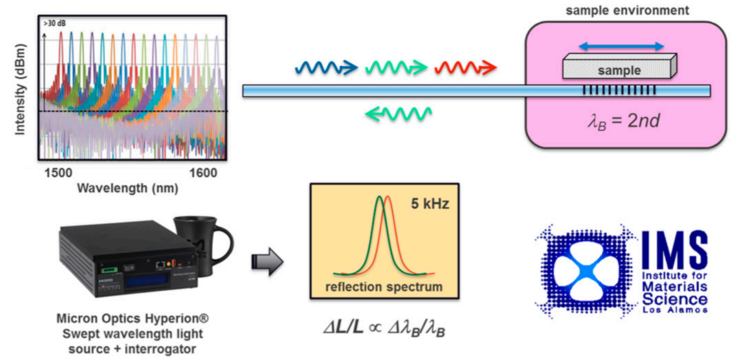
continued on next page

Novel sensor techniques cont.

gies that reduce strain-lag over a broader range of sample geometries.

The Los Alamos portion of the research was sponsored by the National High Magnetic Field Laboratory (NHMFL) through the NHMFL User Collaboration Grant Program, the DOE BES “Science at 100T” program, and the LANL Institute for Materials Science. The Lab contributed oriented single crystal samples over a period of several years for this study, which supports its Materials for the Future and Science of Signatures science pillars by creating effective, science-based strategies to observe and exploit properties of materials. These strategies may open the way to discovering new crystal structure-related signatures of materials under extremes.

Researchers: Marcelo Jaime (National High Magnetic Field Laboratory, NHMFL-PFF and Institute for Materials Science, Los Alamos), Carolina Corvalá n Moya (NHMFL-PFF; Comisión Nacional de Energia Atómica, Argentina; Consejo Nacional de Investigaciones Científicas y Técnicas, Argentina; Universidad Nacional Tres de Febrero, Argentina), Franziska Weickert (formerly NHMFL at Florida State University, Tallahassee, now Materials Synthesis and Integrated Devices, MPA-11), Vivien Zapf and Fedor F. Balakirev (NHMFL-PFF), Mark Wartenbe (NHMFL), Priscila



An experimental setup for the FBG experiment using a swept wavelength laser source. λ_B is the Bragg wavelength, *n* is the laser source, and *d* is the grating spacing.

F. S. Rosa (Condensed Matter and Magnet Science, MPA-CMMS), Jonathan B. Betts (NHMFL-PFF), George Rodriguez (Center for Integrated Nanotechnologies, MPA-CINT), Scott A. Crooker (NHMFL-PFF), and Ramzy Daou (Centre National de la Recherche Scientifique, France).

Reference: “Fiber Bragg grating dilatometry in extreme magnetic field and cryogenic conditions,” *Sensors* **17,** 2572 (2017).

Technical contact: Marcelo Jaime

HeadsUP!

Use bear-proof dumpsters around the site ... correctly.

Purchased by Maintenance and Site Services, bear-proof dumpsters are designed to keep bears out, therefore reducing their motivation to come on site for a free meal.

Keep bears out by ensuring that

- both sets of dumpster doors are shut,
- the lock bar is slid fully into place, and
- all carabiners are in place and clipped.

BEAR-PROOF

Celebrating service

Congratulations to the following MPA Division employees celebrating recent service anniversaries:

Jason Lashley, MPA-11 .... 25 years
Eric Davis, MPA-11 .... 5 years
Ulises Martinez, MPA-11 .... 5 years

MPA Materials Matter

Materials Physics and Applications

Published by the Experimental Physical Sciences Directorate

To submit news items or for more information, contact Karen Kippen, ADEPS Communications, at 505-606-1822 or adeps-comm@lanl.gov. To read past issues see www.lanl.gov/orgs/mpa/materialsmatter.shtml.

Los Alamos NATIONAL LABORATORY EST. 1943

NNSA National Nuclear Security Administration